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Experimental study of flammability limits of natural gas-air mixture

S.Y. Liao^a, Q. Cheng^b, D.M. Jiang^{a,*}, J. Gao^a

^a School of Energy and Power Engineering, Xi'an Jiaotong University, Xi'an 710049, PR China
^b Chongqing Communication Institute, Chongqing 400035, PR China

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Abstract

Flammability limits data are essential for a quantitative risk assessment of explosion hazard associated with the use of combustible gas. The present work is to obtain the fundamental flammability data for prevention of the hazards in the practical applications. Experiments have been conducted in a constant volume combustion bomb, and the fuel considered here is natural gas (NG). The pressure histories in the combustion bomb are recorded and a criterion of 7% pressure rise has been used to judge a flammable mixture. The effects of ethane on NG-air flammability limits have been investigated. By adding diluent (carbon dioxide, nitrogen or their mixture) into NG-air mixture, the dilution effects on the flammability limits have been explored as well, and the results are plotted as functions of diluent ratio. © 2004 Elsevier B.V. All rights reserved.

Keywords: Natural gas; Flammability limit; Diluent addition; Explosion hazard; Combustion bomb

1. Introduction

Knowledge of the explosion hazards of gaseous fuels is of importance to ensure the safety in industrial and domestic applications that produce or use flammable mixtures. There is no question that the flammability limit is a most widely used index for representing the flammability characteristics of gases. In accordance with generally accepted usage, the flammability limits are known as those regions of fuel–air ratio within which flame propagation can be possible and beyond which flame cannot propagate. And there are two distinct separate flammability limits for the fuel–air mixture, namely, the leanest fuel-limit up to which the flame can propagate is termed as lower flammability limit (LFL), and the richest limit is called as upper flammability limit (UFL).

Flammability limits have been discussed extensively in the combustion literature. There are several criteria to determine the flammability limits. A successful attempt can be determined by one or a combination of the following criteria: (1)

inspection of the visualization of the flame kernel produced by the spark, namely visual criterion, and (2) measurements of pressure or temperature histories in the vessel and appropriate pressure or temperature rise criteria can be used to designate flammability rather than the purely visual observation of flame development. As we know, a successful ignition would induce a rapid pressure increase and temperature rise within a short time, as well as produce a propagating flame front that could be readily observed. Previous gas flammability limit data were obtained mainly in flammability tubes, in those tests, a gas mixture in a vertical tube was ignited and flame propagation was inspected by a visual criterion [1,2]. The wall quenching has a significant effect on the flammability measurement in flammability tube. The larger size of combustion charmer can minimize wall effects and can allow for the potential use of stronger igniters to ensure the absence of ignition limitations, so most of the flammability measurements are conducted in closed chambers recently [3–7]. And more attentions are being given to the effects of environmental parameters, such as the vessel size, initial temperature and pressure on the fundamental characteristics. Moreover, the theoretical studies are carried out for providing analytical predictions about the flammability limits [8–11]. With the

^{*} Corresponding author. Tel.: +86 29 82663421; fax: +86 29 2668789. *E-mail address:* dmjiang@mail.xjtu.edu.cn (D.M. Jiang).

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growing crisis of energy resources and the strengthening of pollutant legislations, the use of natural gas (NG) as an alternative fuel has been promoted recently, natural gas is being regarded as one of the most promising alternative fuels for industrial and domestic applications. The chemical composition of natural gas varies from field to field, but the main chemical component of natural gas is believed to be methane. There is a large volume of flammability limits data available for fuels, such as methane, ethane, propane, butane, etc. But to the best of our knowledge, no work has been reported so far on the flammability limits of NG-air mixture. Therefore, the present work is promoted purposely. The experiments are made systematically to determine the flammability limits of premixed NG-air mixture in a constant volume combustion bomb, using conventional spark ignition system. The explosion pressure traces are recorded and an appropriate pressure rise criterion is used to define the flammability limit. The natural gas selected for the present study is from the north of Shannxi province, PR China, which consists of 96.160% methane by volume, 2.540% carbon dioxide, 1.096% ethane, approximately 0.189% hydrocarbon components higher than C3, and the remains including nitrogen, sulfureted hydrogen and water are only about 0.015%.

The dilution effects on the fundamental characteristic data of natural gas are also studied by adding the carbon dioxide, nitrogen gas and the simulated exhaust gas into the NG–air mixture, where the simulated gas contains 12% CO₂ and 88% N₂. And the results are plotted as functions of diluent ratio, where the diluent ratio ϕ_{γ} denotes the volumetric fraction of dilution addition in the total mixture.

2. Experimental method

The experimental investigation is conducted in a constant volume combustion bomb, as shown in Fig. 1. The cubic combustion bomb has an inside size of $108 \text{ mm} \times 108 \text{ mm} \times 135 \text{ mm}$, with 1.571 in volume. Two sides of this bomb are fixed quartz glasses to make the inside observable, which are to provide the viewing access for the observation of flame growth. The combustible mixture is prepared within the closed vessel by adding gases needed to appropriate partial pressures. Two extended stainless steel electrodes are used to form the spark gap at the center of this bomb. Center ignition in the combustion bomb is attempted using 45 mJ igniter, stored in a conventional battery–coil ignition system. Dynamic pressure histories are measured since spark ignition with an absolute pressure transducer, model Kistler 4075A, with a calibrating element Kistler 4618A.

Following the standard test procedures developed by the Pittsburgh Research Laboratory (PRL) and according to the previous experiment [6], a criterion of 7% pressure rise is used to distinguish flammable mixtures from nonflammable ones. When a pressure increase of 7% is recorded after ignition, i.e. 7% above the initial absolute pressure, it is indicative of that the occurrence of combustion is successful.



Fig. 1. Schematic diagram of experimental setup.

3. Results and discussions

The data plotted in Fig. 2 is the result of tests on the quiescent NG–air mixtures at ambient pressure and temperature. It plots the maximum explosion pressure as function of the NG concentration. The baseline of critical pressure rise rate, 1.07, is plotted as well, thereby, one can readily observe that the flammability region of quiescent NG–air mixtures is from 5.0 to 15.6% of NG by volume, corresponding to the equivalence ratios from about 0.486 to 1.707.

In order to validate our experimental measurements, a comparison is made with the available flammability limit data for methane–air mixture, as shown in Table 1. The present result is in agreement with those of previous works even though the test conditions are different. As to the lower flammability limit of multiple component fuel mixture in air, the LFL value of the mixture can also be estimated by following formula,

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Flammability limit data (vol.%) for methane-air and NG-air flames (quiescent mixtures, with spark ignition)

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Mixtures	Test conditions	LFL (vol.%)	UFL (vol.%)
NG–air ^a	1.57 L chamber	5.0	15.6
	LeChatelier's rule	4.98	-
Methane-air 8 L chamb 20 L cham 120 L cham 25.5 m ³ sp Flammabi	8 L chamber ^b	5.0	_
	20 L chamber ^c	4.9	15.9
	120 L chamber ^d	5.0	15.7
	25.5 m ³ sphere	$4.9^{\rm e}; 5.1 \pm 0.1^{\rm f}$	_
	Flammability tubeg	4.9	15.0

^a The present study, using pressure rise criterion of 7%.

^b Hertzberg and Cashdollar (1983), using pressure rise criterion of 7%, from [3].

^c Cashdollar et al. (2000), using pressure rise criterion of 7% [3].

^d Cashdollar et al. (2000), using pressure rise criterion of 7% [3].

^e Burgess et al. (1982), using a visual criterion, from [3].

^f Furno et al. (1970) and Burgess et al. (1982), using pressure rise criterion of 7%, from [3].

^g Kuchta (1985), using a visual criterion [8].



Fig. 2. Determinations of the flammability limits of NG–air mixtures, where hollow point represents a successful combustion and solid one is an unsuccessful attempt: (A) the determination of LFL, and (B) the determination of UFL.

according to LeChatelier's rule [6]:

$$LFL = \frac{100}{\sum (C_i/LFL_i)}$$
(1)

where LFL is the lower flammability limit of mixture (vol.%), C_i the concentration of component *i* in the gas mixture on an air-free basis (vol.%), and LFL_i is the lower flammability limit for component i (vol.%). The estimation of LeChatelier's rule is listed in Table 1 as well. Generally, good agreement has been found between the measurement and LeChatelier prediction. The effects of ethane on flammability limits are explored by adding ethane into the natural gas mixture. The results are presented in Fig. 3. It is reported that the flammability region has been slightly extended with the increase of ethane content in natural gas. Over the studied ranges, the value of LFL almost remains 5.0 in volume, but UFL has become bigger. The flammability limits are 3.0 to 12.5 in volume for ethane-air mixture [8], corresponding to equivalence ratios of 0.512 and 2.506, however, as to methane, they are 0.486 to 1.707, respectively. Hereby, the increase of ethane content in NG can also extend the UFL in



Fig. 3. Dependence of NG flammability limits on ethane.

equivalence ratio, whereas, there is no obvious change for LFL in the equivalence ratio. Similarly, the LFL values obtained from LeChatelier's prediction are presented in Fig. 3.

Nowadays, the method of exhaust gas recirculation has being widely used for it has being regarded as an effective way to decrease NO_x emissions in combustion engine [12]. Experimental measurements are performed to investigate the effects on the flammability limits of diluting NG-air mixture by adding carbon dioxide, nitrogen gas and the simulated combustion products. The dilutions used to simulate combustion products consist of 88% N2 and 12% CO2 by volume in this work, which is in reasonably agreement with the practical products for NG stoichiometric combustion at ambient temperature. Shown in Fig. 4, are the results of flammability limits over a wide range of diluent ratio. It can be noted that the increase of diluent ratio results in the lessening of flammability region. The reason is known as the addition of dilutions decreases the temperature of flame, consequently decreases the burning velocity. Hence, the flammability limits becomes narrow. Generally, CO2 addition is much more sensitive than N2 addition [13], thus, there is much difference between additive N₂ and CO₂. Shebeko et al. [11] proposed an analytical evaluation of flammability limits for ternary gaseous mixtures of fuel-air-diluent. The predictions are presented in Fig. 4



Fig. 4. Flammability limits of diluting NG-air mixtures, where dashed curves are the predictions based on Shebeko method [11].

with dashed curves as well. It must be noted that the general trends are obtained from the predictions and the derivations between the measurements and they are acceptable.

4. Conclusions

This paper is to determine the flammability limit data of natural gas–air mixture. Experimental investigations are conducted by systematically measuring the pressure histories in a constant volume combustion bomb. It is reported that the flammability region is from 5.0% to 15.6% of NG by volume at ambient conditions, for quiescent NG–air mixture with spark igniter, which is comparable with that of pure methane in air. The effects of ethane and diluent addition on the flammability limits are explored experimentally as well. Moreover, LeChatelier's rule and Sheboke analytical method have been used to predict the flammability characteristics, and from the comparable results obtained, our measurement has been justified.

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